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Atty's Docket No. PET-1845

U.S. PTO  
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Applicant(s): **Dominique COMMEREUC ET AL.**

**IMPROVED CATALYTIC COMPOSITION AND ITS APPLICATION TO OLEFIN  
OLIGOMERIZATION**

THE COMMISSIONER OF PATENTS & TRADEMARKS  
Washington, D.C. 20231

Sir:

Herewith is the above-identified application for Letters Patent including:

- ☒ Specification and claims ☐ Verified statement(s) to establish small entity status under 37 CFR 1.9 and 37 CFR 1.27
- ☐ \_\_\_\_\_ Sheets Drawings
- ☐ Formal ☐ Informal ☒ Information Disclosure
- ☒ Declaration and Power of Attorney ☐
- ☒ Preliminary Amendment
- ☒ A check in the amount of \$ 690.00 is attached.
- ☐ Please charge my Deposit Account No. 13-3402 in the amount of \$ \_\_\_\_\_  
2 copies of this sheet are attached.

CLAIMS AS FILED					
	FOR	NUMBER FILED	NUMBER EXTRA	RATE	BASIC FEE \$690.00
	TOTAL CLAIMS	20 -20 =	0	x	0.00
	INDEPENDENT CLAIMS	1 - 3 =	0	x	0.00
	<input type="checkbox"/> Multiple Dependent Claim Presented				
			TOTAL FILING FEE		\$690.00

- ☒ The benefit under 35 USC 119 is claimed of the filing date of:  
**FRENCH APPLICATION NO. 99/06.749, filed May 27, 1999**
- ☒ A certified copy of the priority document(s) is attached.
- ☒ The Commissioner is hereby authorized to charge any deficiencies in payment of the following fees associated with this communication or credit any overpayment to Deposit Account No. 13-3402.
- ☒ Any filing fees under 37 CFR 1.16 for the presentation of extra claims.
- ☒ Any patent application processing fees under 37 CFR 1.17.
- ☒ The Commissioner is hereby authorized to charge payment of the following fees during the pendency of this application or credit any overpayments to Deposit Account No. 13-3402, two copies of this sheet are being enclosed.
- ☒ Any patent application processing fees under 37 CFR 1.17
- ☐ The issue fee set in 37 CFR 1.18 at or before mailing of the Notice of Allowance, pursuant to 37 CFR 1.311(b).
- ☒ Any filing fees under 37 CFR 1.16 for presentation of extra claims

Respectfully submitted,

**MILLEN, WHITE, ZELANO & BRANIGAN, P.C.**

BY: *William Millen*  
**I. William Millen (19,544)**  
Attorney for Applicants

DATE: May 26, 2000

**IN THE UNITED STATES PATENT AND TRADEMARK OFFICE**

In re application of : **BOX PATENT APPLICATION**

Dominique COMMEREUC et al. : Examiner: Unassigned

Serial No.: Unassigned : Group Art Unit: Unassigned

Filed: May 26, 2000 :

For: IMPROVED CATALYTIC COMPOSITION AND ITS APPLICATION TO OLEFIN  
OLIGOMERIZATION

**PRELIMINARY AMENDMENT**

Assistant Commissioner for Patents  
Washington, D.C. 20231

Sir:

Prior to examination, Applicants wish to amend the above-identified application as indicated below:

**IN THE CLAIMS:**

Please amend the claims as follows:

**Claim 1, Line 1:** Delete "characterized in that it comprises the" and insert --comprising a--.

**Claim 2, Line 1:** Delete "characterized in that" and insert --wherein--.

**Claim 3, Line 1:** Delete "or claim 2, characterized in that" and insert --wherein--.

**Line 2:** Delete "selected from the group formed by" and insert --a--.

**Line 3:** Delete "compounds with" and insert --of the--.

**Line 4:** Delete ", in particular those which contain at least one halogen atom alpha to the -COOH" and insert --.--.

**Line 5:** Delete in its entirety.

- Claim 4, Line 1:** Delete “any one of claims 1 to 3, characterized in that” and insert --claim 3, wherein--.
- Claim 5, Line 1:** Delete “any one of claims 1 to 4, characterized in that” and insert --claim 4, wherein--.
- Claim 6, Line 1:** Delete “any one of claims 1 to 5, characterized in that” and insert --claim 1, wherein--.
- Line 2:** Delete “consists of” and insert --comprises--.
- Claim 7, Line 1:** Delete “any one of claims 1 to 6, characterized in that” and insert --claim 1, wherein--.
- Claim 8, Line 1:** Delete “any one of claims 1 to 7, characterized in that” and insert --claim 1, wherein--.
- Claim 9, Line 1:** Delete “characterized in that” and insert --wherein--.
- Claim 10, Line 1:** Delete “or claim 9, characterized in that” and insert --wherein--.
- Line 3:** After “AIX<sub>3</sub>” delete “,” and insert --.--.
- Line 4:** Delete in its entirety.
- Claim 11, Line 1:** Delete “or claim 9, characterized in that” and insert --wherein--.

**12. (Amended)** A process for dimerization or oligomerization of at least one monoolefin, [characterized in that] comprising contacting said monoolefin [is brought into contact] with a catalytic composition according to [any one of claims 1 to 11] claim 1.

**13. (Amended)** A process according to claim 12, [characterized in that] wherein the pre-conditioning solvent for the catalytic composition [consists of] comprises a mixture of olefins [with] having a composition [analogous to] approximating that of the mixtures obtained by [the] said dimerization or oligomerization reaction.

**Claim 14,**     **Line 1:**        Delete “or claim 13”.

**Line 2:**        Delete “characterized in that” and insert --wherein--.

**Please add the following claims:**

--     **15.**     A process according to claim 12, wherein the pre-conditioning is conducted with stirring under an inert atmosphere at 0°C to 80°C for 1 minute to 5 hours, and the catalyst is then transferred to a reactor under an inert atmosphere.

**16.**     A process according to claim 12, wherein the preconditioning is conducted with stirring under an inert atmosphere at 10° to 60° for 5 minutes to 1 hour, and the catalyst is then transferred to a reactor under an inert atmosphere.

**17.**     A catalyst composition according to claim 3, wherein the halogenocarboxylic acid has a total of 2 to 20 carbon atoms and contains at least one halogen atom alpha to the -COOH group.

**18.**     A catalyst composition according to claim 1, being devoid of ethylene, propylene and butene.

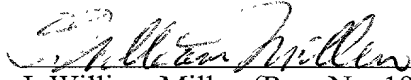
**19.**     A catalytic composition according to claim 18, wherein pre-conditioning comprises mixing the three constituents in a hydrocarbon or halogeno-hydrocarbon solvent with stirring and in an inert atmosphere at a controlled temperature of 0°C to 80°C and for a duration of 1 minute to 5 hours.

**20.**     A catalyst composition according to claim 19, wherein the pre-conditioning solvent comprises isohexenes.--

**REMARKS**

A principal purpose of this Preliminary Amendment is to remove the multiply dependent claims and avoid the fee associated therewith, applicant reserving the right to reintroduce claims to canceled combined subject matter.

Respectfully submitted,



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**Filed: May 26, 2000**

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# IMPROVED CATALYTIC COMPOSITION AND ITS APPLICATION TO OLEFIN OLIGOMERIZATION

## Field of the Invention

The present invention relates to a novel catalytic composition for use in oligomerization  
5 processes, in particular for monoolefin dimerization.

It also relates to a process for oligomerization, in particular dimerization, of monoolefins,  
using such a catalytic composition.

## Description of the Prior Art

0093350362103550  
10 The preparation of catalysts for dimerization or co-dimerization of monoolefins such as  
ethylene, propylene, butenes or pentenes is known. Examples of such catalysts which can be cited  
are the products of the interaction of  $\pi$ -allyl nickel phosphine halides with Lewis acids (French  
patent FR-B-1 410 430), the products of the interaction of nickel phosphine halides with Lewis acids  
(United States patent US-A-3 485 881) and the products of the interaction of certain nickel  
carboxylates with hydrocarbylaluminium halides (US-A-3 321 546).

15 Almost all of those catalysts use a ligand such as an organic phosphorus compound and it is  
preferable to have phosphorus-free oligomerization catalysts available. To this end, it is possible to  
use zerovalent nickel compounds but they are of little practical use because of their instability. It is  
also possible to use catalysts in which the nickel is deposited on a mineral support comprising acid  
sites, such as silica, alumina or silica-aluminas. However, in that case the catalysts are solid, in  
20 contrast to the desired liquid catalysts.

US-A-4 283 305 teaches that an association of a divalent nickel compound, a hydrocarbyl-  
aluminium halide with formula  $AlR_mX_{3-m}$  where R is a hydrocarbyl radical containing 1 to 12 carbon  
atoms, X is a chlorine or bromine atom, and m is a number from 1 to 2, and a compound with a  
Bronsted acid nature, leads to a catalytic composition that is more active than previously, and also  
25 less sensitive to the trace impurities which are routinely found in industrial olefinic feeds.

## Summary of the Invention

It has now, unexpectedly, been found that for a catalytic composition obtained by bringing a  
divalent nickel compound into contact with an organic Bronsted acid and a hydrocarbylaluminium

dihalide, pre-conditioning the catalytic composition in a solvent before using it for oligomerization can still further increase the activity for olefin oligomerization. The use of a hydrocarbylaluminium dihalide enriched with an aluminium trihalide can still further increase the activity of the catalytic composition.

## 5 Detailed Description of the Invention

More precisely, said improved catalytic composition comprises the product resulting from bringing the following three constituents into contact in any order:

- a) at least one divalent nickel compound;
- b) at least one hydrocarbylaluminium dihalide with formula  $AlRX_2$ , where R is a hydrocarbyl radical containing 1 to 12 carbon atoms such as alkyl, aryl, aralkyl or cycloalkyl, and X is a chlorine or bromine atom; and
- c) at least one organic Bronsted acid;

the mixture obtained being pre-conditioned in a solvent, at a controlled temperature and for a pre-set period, prior to its use.

The divalent nickel compound can be any compound soluble in a proportion of more than 1 g per litre in a hydrocarbon medium, more particular in the reactants and the reaction medium. Preferably, nickel carboxylates are used with general formula  $(R_1COO)_2Ni$ , where  $R_1$  is a hydrocarbyl radical, for example alkyl, cycloalkyl, alkenyl, aryl, aralkyl or alkaryl, containing up to 20 carbon atoms, preferably a hydrocarbyl residue containing 5 to 20 carbon atoms. Radical  $R_1$  can be substituted by one or more halogen atoms, hydroxyl groups, ketone, nitro, cyano or other groups that do not interfere with the reaction. The two radicals  $R_1$  can also constitute an alkylene residue containing 6 to 18 carbon atoms. Non limiting examples of nickel compounds are the following divalent nickel salts: octoate, 2-ethylhexanoate, decanoate, stearate, oleate, salicylate and hydroxy-decanoate. Preferably, nickel 2-ethylhexanoate is used.

The Bronsted acid compound has formula  $HY$ , where Y is an organic anion, for example carboxylic, sulphonic or phenolic. Preferably, acids with a  $pK_a$  of a maximum of 3 at 20°C are used, more particularly those which are soluble in the nickel compound or in its solution in a hydrocarbon or another suitable solvent. One preferred class of acids comprises the group formed by

halogenocarboxylic compounds with formula  $R_2COOH$  where  $R_2$  is a halogenated alkyl radical, in particular those which contain at least one halogen atom alpha to the  $-COOH$  group, with a total of 2 to 10 carbon atoms. Preferably, a halogenoacetic acid with formula  $CX_pH_{3-p}-COOH$  is used where X is fluorine, chlorine, bromine or iodine, and p is a whole number from 1 to 3. Examples which can be cited are trifluoroacetic, difluoroacetic, fluoroacetic, trichloroacetic, dichloroacetic and chloroacetic acids. These examples are not limiting and it is also possible to use arylsulphonic acids, alkylsulphonic acids, fluoroalkylsulphonic acids, picric acid and nitroacetic acid. Preferably, trifluoroacetic acid is used.

The three constituents of the catalytic formula can be mixed in any order. However, it is preferable to first mix the nickel compound with the organic Bronsted acid then to introduce the aluminium compound. The mole ratio of the hydrocarbylaluminium dihalide to the nickel compound, expressed as the Al/Ni ratio, is 2/1 to 50/1, preferably 2/1 to 20/1. The mole ratio of the Bronsted acid to the nickel compound is 0.25/1 to 10/1, preferably 0.25/1 to 5/1.

Pre-conditioning the catalytic composition consists of mixing the three constituents in a hydrocarbon solvent, for example in an alkane or in an aromatic hydrocarbon, or in a halogenated hydrocarbon or, as is preferred, in a mixture with a composition analogous to that of the mixtures obtained in the dimerization or oligomerization reaction itself. Thus for a catalytic composition intended for propylene dimerization, the pre-conditioning solvent can principally be constituted by isohexenes.

The mixture is generally produced by stirring in an inert atmosphere, for example in nitrogen or argon, at a controlled temperature of  $0^{\circ}C$  to  $80^{\circ}C$ , preferably  $10^{\circ}C$  to  $60^{\circ}C$ , for a period of 1 minute to 5 hours, preferably 5 minutes to 1 hour. The solution obtained is then transferred into the oligomerization reactor under an inert atmosphere.

In a preferred implementation, in the catalytic composition of the invention, the hydrocarbylaluminium dihalide can be enriched with an aluminium trihalide, the mixture of the two compounds then having formula  $AlR_nX_{3-n}$ , where R and X are as defined above and n is a number from 0 to 1 (limits excluded).



The hydrocarbylaluminium dihalide compounds enriched in aluminium trihalide are obtained by mixing a hydrocarbylaluminium dihalide with formula  $\text{AlRX}_2$  where R is a hydrocarbyl radical containing 1 to 12 carbon atoms, such as alkyl, aryl, aralkyl, alkaryl or cycloalkyl and X is a chlorine atom or a bromine atom, with an aluminium trihalide  $\text{AlX}_3$ . Non limiting examples of such compounds which can be cited are: dichloroethylaluminium enriched with aluminium trichloride, the mixture having formula  $\text{AlEt}_{0.9}\text{Cl}_{2.1}$ , for example; dichloroisobutylaluminium enriched with aluminium trichloride, the mixture having formula  $\text{AliBu}_{0.9}\text{Cl}_{2.1}$ , for example, and dibromoethylaluminium enriched with aluminium tribromide, the mixture having formula  $\text{AlEt}_{0.9}\text{Br}_{2.1}$ , for example.

In this case as well, the three constituents of the catalytic formula can be mixed in any order. It is also preferable to first mix the nickel compound with the organic Bronsted acid, then to introduce the aluminium compound. In this case, it is = the mole ratio between the hydrocarbylaluminium dihalide enriched with aluminium trihalide and the nickel compound, expressed as the Al/Ni ratio, which is 2/1 to 50/1, preferably 2/1 to 20/1. As indicated above, the mole ratio of the Bronsted acid to the nickel compound is still 0.25/1 to 10/1, preferably 0.25/1 to 5/1.

The invention also relates to a process for oligomerization, in particular dimerization, of monoolefins in the presence of the catalytic system defined above.

Examples of monoolefins that can be dimerized or oligomerized are ethylene, propylene, butenes, pentenes and hexenes, used pure or as a mixture, contained in cuts from refining or from chemistry. These olefins can also be co-oligomerized between themselves.

The process can be carried out in a reactor with one or more reaction stages in series, the olefinic feed and/or the previously pre-conditioned catalytic composition being introduced continuously, either to the first stage, or to the first and to any other stage.

The process is generally carried out at a temperature of  $-20^\circ\text{C}$  to  $+80^\circ\text{C}$ , under pressure conditions such that at least the major portion of the reactants are maintained in the liquid phase or in the condensed phase.

At the reactor outlet, the catalyst can be deactivated, for example by injecting ammonia and/or an aqueous sodium hydroxide solution and/or an aqueous sulphuric acid solution. The

unconverted olefins and alkanes which may be present in the feed are then separated from the oligomers by distillation.

The products obtained by the process of the invention can be used, for example, as constituents for automobile fuels and/or as feeds for a hydroformylation process for synthesising aldehydes and alcohols.

The entire disclosure of all applications, patents and publications, cited above and below, and of corresponding French application 99/06749, filed on May 27, 1999 are hereby incorporated by reference.

The following examples illustrate the invention without in any way limiting its scope.

#### **EXAMPLE 1 (comparative)**

##### Preparation of catalyst:

In this example of the prior art, the catalyst was prepared in situ in the autoclave where oligomerization took place, without pre-conditioning, and used dichloroethylaluminium as the aluminium compound.

##### Use in oligomerization:

A solution of 0.043 g of nickel 2-ethylhexanoate containing 13% by weight of nickel in 40 ml of isohexene solvent was introduced into a 250 ml stainless steel autoclave provided with stirring and wherein the temperature could be regulated by circulating water in an external envelope, followed by 10 g of a mother liquor prepared from 0.11 g of trifluoroacetic acid made up to 100 g with isohexene solvent, introduced with stirring. Finally, a solution of 0.18 g of dichloroethylaluminium in 50 ml of isohexene solvent was injected. This corresponded to 1.41 mmole of aluminium and an Al/Ni mole ratio of 15/1.

Subsequently, 10 g of isohexenes was added to the autoclave to make up the solvent, followed by 10.8 g of liquid propylene via pressure lock. The temperature was rapidly raised to 40°C. After 15 minutes of reaction, the conversion of propylene into a mixture of dimers, trimers and tetramers containing 80% by weight of dimers was 67%.

#### **EXAMPLE 2 (in accordance with the invention)**

##### Preparation of catalyst:

0.043 g of nickel 2-ethylhexanoate containing 13% by weight of nickel was introduced into a 250 ml glass flask provided with a magnetic stirrer, then the flask was carefully purged and placed under an argon atmosphere. A transfer needle was used to introduce 40 ml of a fraction of isohexenes distilled under argon and dried over 3A molecular sieve, which was then used as the solvent. Stirring dissolved the nickel salt. 10 g of a mother liquor prepared from 0.11 g of trifluoroacetic acid made up to 100 g with the isohexene solvent was then injected. This was all placed, with continued stirring, in a thermostatic bath regulated to 30°C.

A further flask purged with argon was used to prepare a solution of 0.18 g of dichloroethylaluminium in 50 ml of isohexene solvent. The solution obtained was slowly added to the nickel solution prepared above using a transfer needle. This corresponded to 1.41 mmole of total aluminium and to a Al/Ni mole ratio of 15/1. This was all pre-conditioned at 30°C for 30 minutes with stirring.

Use in oligomerization:

The pre-conditioned catalytic solution was transferred under argon to an autoclave as described in Example 1. 10 g of isohexenes was then introduced into the autoclave to make up the solvent, followed by 10.8 g of liquid propylene using a pressure lock. The temperature was rapidly raised to 40°C. The reaction was followed by periodically removing samples for gas chromatographic analysis. After 15 minutes of reaction, the conversion of propylene into a mixture of dimers, trimers and tetramers analogous to that of Example 1 was 86%.

**EXAMPLE 3 (in accordance with the invention)**

Preparation of catalyst:

0.043 g of nickel 2-ethylhexanoate containing 13% by weight of nickel was introduced into a 250 ml glass flask provided with a magnetic stirrer, then the flask was carefully purged and placed under an argon atmosphere. A transfer needle was used to introduce 40 ml of a fraction of isohexenes distilled under argon and dried over 3A molecular sieve, which was then used as the solvent. Stirring dissolved the nickel salt. 10 g of a mother liquor prepared from 0.11 g of trifluoroacetic acid made up to 100 g with the isohexene solvent was then injected. This was all placed, with continued stirring, in a thermostatic bath regulated to 30°C.

50 ml of isohexene solvent was introduced into a further flask purged with argon, followed by 0.165 g of dichloroethylaluminium and finally 0.015 g of aluminium trichloride, corresponding to a compound with formula  $\text{AlEt}_{0.92}\text{Cl}_{2.08}$ . The solution obtained was slowly added to the nickel solution prepared above using a transfer needle, whereupon the colour changed from green to yellow. This corresponded to 1.41 mmole of total aluminium and to an Al/Ni mole ratio of 15/1. This was all pre-conditioned at 30°C for 30 minutes with stirring.

Use in oligomerization:

The pre-conditioned catalytic solution was transferred under argon to an autoclave as described in Example 1. 10 g of isohexenes was then introduced into the autoclave to make up the solvent, then 10.8 g of liquid propylene using a pressure lock. The temperature was rapidly raised to 40°C. The reaction was followed by periodically removing samples for gas chromatographic analysis. After 15 minutes of reaction, the conversion of propylene into a mixture of dimers, trimers and tetramers analogous to that of Example 1 was 89%.

The preceding examples can be repeated with similar success by substituting the generically or specifically described reactants and/or operating conditions of this invention for those used in the preceding examples.

From the foregoing description, one skilled in the art can easily ascertain the essential characteristics of this invention and, without departing from the spirit and scope thereof, can make various changes and modifications of the invention to adapt it to various usages and conditions.

## CLAIMS

1. A catalytic composition, characterized in that it comprises the product resulting from bringing the following three constituents into contact in any order:
  - a) at least one divalent nickel compound;
  - 5 b) at least one hydrocarbylaluminium dihalide with formula  $\text{AlRX}_2$ , where R is a hydrocarbyl radical containing 1 to 12 carbon atoms such as alkyl, aryl, aralkyl or cycloalkyl, and X is a chlorine or bromine atom; and
  - c) at least one organic Bronsted acid;

the mixture obtained being pre-conditioned in a solvent, at a controlled temperature and for a pre-set period, prior to its use.
- 10 2. A catalytic composition according to claim 1, characterized in that said divalent nickel compound is a nickel carboxylate with general formula:
 
$$(\text{R}_1\text{COO})_2\text{Ni}$$

where  $\text{R}_1$  is an alkyl, cycloalkyl, alkenyl, aryl, aralkyl or alkaryl radical containing up to 20 carbon atoms.
- 15 3. A catalytic composition according to claim 1 or claim 2, characterized in that the  $\text{pK}_a$  of said organic Bronsted acid is a maximum of 3 at  $20^\circ\text{C}$  and is selected from the group formed by halogenocarboxylic compounds with formula  $\text{R}_2\text{COOH}$  where  $\text{R}_2$  is a halogenated alkyl radical, in particular those which contain at least one halogen atom alpha to the  $-\text{COOH}$  group, with a total of 2 to 10 carbon atoms.
- 20 4. A catalytic composition according to any one of claims 1 to 3, characterized in that said organic Bronsted acid is a halogenoacetic acid with formula  $\text{CX}_p\text{H}_{3-p}-\text{COOH}$  is used where X is fluorine, chlorine, bromine or iodine, and p is a whole number from 1 to 3.
5. A catalytic composition according to any one of claims 1 to 4, characterized in that said organic Bronsted acid is trifluoroacetic acid, trichloroacetic acid or tribromoacetic acid.
- 25 6. A catalytic composition according to any one of claims 1 to 5, characterized in that the pre-conditioning consists of mixing the three constituents in a hydrocarbon or halogeno-

hydrocarbon solvent with stirring and in an inert atmosphere at a controlled temperature of 0°C to 80°C and for a duration of 1 minute to 5 hours.

7. A catalytic composition according to any one of claims 1 to 6, characterized in that the mole ratio of said hydrocarbylaluminium dihalide to said nickel compound, expressed as the Al/Ni ratio, is 2/1 to 50/1, and the mole ratio of said Bronsted acid to said nickel compound is 0.25/1 to 10/1.
8. A catalytic composition according to any one of claims 1 to 7, characterized in that said hydrocarbylaluminium dihalide is enriched with an aluminium trihalide, the mixture of these two compounds having formula  $AlR_nX_{3-n}$ , R and X being as defined in claim 1 and where n is a number between 0 and 1.
9. A catalytic composition according to claim 8, characterized in that the mole ratio between said hydrocarbylaluminium dihalide enriched with an aluminium trihalide and the nickel compound, expressed as the ratio Al/Ni, is 2/1 to 50/1, and the mole ratio of the Bronsted acid to the nickel compound is 0.25/1 to 10/1.
10. A catalytic composition according to claim 8 or claim 9, characterized in that said hydrocarbylaluminium dihalide enriched with an aluminium trihalide is obtained by mixing a hydrocarbylaluminium dihalide with formula  $AlRX_2$  with an aluminium trihalide  $AlX_3$ , where R and X are as defined in claim 1.
11. A catalytic composition according to claim 8 or claim 9, characterized in that said hydrocarbylaluminium dihalide enriched with an aluminium trihalide is obtained by mixing dichloroethylaluminium with aluminium trichloride.
12. A process for dimerization or oligomerization of at least one monoolefin, characterized in that said monoolefin is brought into contact with a catalytic composition according to any one of claims 1 to 11.
13. A process according to claim 12, characterized in that the pre-conditioning solvent for the catalytic composition consists of a mixture of olefins with a composition analogous to that of the mixtures obtained by the dimerization or oligomerization reaction.

14. A process according to claim 12 or claim 13, in which the propylene is dimerized or oligomerized, characterized in that the pre-conditioning solvent for the catalytic composition principally comprises isohexenes.

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## IMPROVED CATALYTIC COMPOSITION AND ITS APPLICATION TO OLEFIN OLIGOMERIZATION

### Abstract of the Disclosure:

An improved catalytic composition for oligomerization, in particular dimerization, of monoolefins comprises the product resulting from bringing the following three constituents into contact in any order:

- a) at least one divalent nickel compound;
- b) at least one hydrocarbylaluminium dihalide, optionally enriched with an aluminium trihalide;  
and
- c) at least one organic Bronsted acid;

the catalytic composition being pre-conditioned in a solvent before using it for oligomerization.



Docket No.  
PET 1845

# Declaration and Power of Attorney For Patent Application

## English Language Declaration

As a below named inventor, I hereby declare that :

My residence, post office address and citizenship are as stated below next to my name,

I believe I am the original, first and sole inventor (if only one name is listed below) or an original, first and joint inventor (if plural names are listed below) of the subject matter which is claimed and for which a patent is sought on the invention entitled

IMPROVED CATALYTIC COMPOSITION AND ITS APPLICATION TO OLEFIN OLIGOMERIZATION

the specification of which

(check one)

☒ is attached hereto.

☐ was filed on

as United States Application No. or PCT international

Application Number

and was amended on

(if applicable)

I hereby state that I have reviewed and understand the contents of the above identified specification, including the claims, as amended by any amendment referred to above.

I acknowledge the duty to disclose to the United States Patent and Trademark Office all information known to me to be material to patentability as defined in Title 37, Code of Federal Regulations, Section 1.56.

I hereby claim foreign priority benefits under Title 35, United States Code, Section 119(a)-(d) or Section 365(b) of any foreign application(s) for patent or inventor's certificate, or Section 365(a) of any PCT International application which designated at least one country other than the United States, listed below and have also identified below, by checking the box, any foreign application for patent or inventor's certificate or PCT International application having a filing date before that of the application on which priority is claimed.

Prior Foreign Application(s)

Priority Not Claimed

99/06.749	France	27/05/99	<input type="checkbox"/>
(Number)	(Country)	(Day/Month/Year Filed)	
_____	_____	_____	<input type="checkbox"/>
(Number)	(Country)	(Day/Month/Year Filed)	
_____	_____	_____	<input type="checkbox"/>
(Number)	(Country)	(Day/Month/Year Filed)	

00550179 052600

I hereby claim the benefit under 35 U.S.C. Section 119(e) of any United States provisional application(s) listed below :

\_\_\_\_\_  
(Application Serial No.)

\_\_\_\_\_  
(Filing Date)

\_\_\_\_\_  
(Application Serial No.)

\_\_\_\_\_  
(Filing Date)

\_\_\_\_\_  
(Application Serial No.)

\_\_\_\_\_  
(Filing Date)

I hereby claim the benefit under 35 U. S. C. Section 120 of any United States application(s), or Section 365(c) of any PCT International application designating the United States, listed below and, insofar as the subject matter of each of the claims of this application is not disclosed in the prior United States or PCT International application in the manner provided by the first paragraph of 35 U.S.C. Section 112. I acknowledge the duty to disclose to the United States Patent and Trademark Office all information known to me to be material to patentability as defined in Title 37, C.F.R., Section 1.56 which became available between the filing date of the prior application and the national or PCT International filing date of this application :

\_\_\_\_\_  
(Application Serial No.)

\_\_\_\_\_  
(Filing Date)

\_\_\_\_\_  
(Status)  
(patented, pending, abandoned)

\_\_\_\_\_  
(Application Serial No.)

\_\_\_\_\_  
(Filing Date)

\_\_\_\_\_  
(Status)  
(patented, pending, abandoned)

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(Application Serial No.)

\_\_\_\_\_  
(Filing Date)

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(Status)  
(patented, pending, abandoned)

I hereby declare that all statements made herein of my own knowledge are true and that all statements made on information and belief are believed to be true ; and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under Section 1001 of Title 18 of the United States Code and that such willful false statements may jeopardize the validity of the application or any patent issued thereon.

POWER OF ATTORNEY : As a named inventor, I hereby appoint the following attorney(s) and/or agent(s) to prosecute this application and transact all business in the Patent and Trademark Office connected therewith. *(list name and registration number)*

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